

Onset of Couette flow in a confined colloidal glass

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Abstract

A colloidal glass confined between rough walls is investigated using non-equilibrium molecular dynamics computer simulations. The onset of flow, under the imposition of a uniform shear stress, is studied. When the imposed stress is gradually decreased, the timescale for the onset of steady flow diverges. Near this yield-stress regime, persistent creep is observed. This regime of slow deformation is shown to be associated with long-lived dynamical heterogeneities, which are strongly influenced by the initial state.

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A defining mechanical property of amorphous solids is the existence of a yield stress; these materials yield only when the applied stress exceeds this threshold [1]. In experiments, the material's response near yielding is commonly probed by using stress as a control parameter [2]. Such measurements report about the existence of a creeping regime, where the material deforms very slowly. Quite often, in such situations, steady flow is not observed in soft amorphous materials (like colloids, gels, emulsions etc.) within experimental durations [3, 4]. This is related to the recent recognition of diverging timescales for the onset of flow [5–7]. Beyond these macroscopic measurements, little is known about the spatio-temporal characteristics of the local dynamics around yielding, which would allow for an improved understanding of the micro-mechanisms at play.

An emerging scenario for the micro-dynamics is that when the material is sheared, structural rearrangements occur locally [8–10] which subsequently trigger more events in the neighbourhood and this correlated process initiates and sustains the flow. Kinetic elastoplastic models [11] based on such a picture of fluidization have been able to explain various steady-state flow properties in confined systems [12–14]. Further, it has been conjectured that the triggering process results in an avalanche-like behaviour, as evidenced via simulations [15, 16]. Here, we focus on how such correlations are expressed when a quiescent glass is subjected to an external stress. The recent measurements near creep [5] seem to indicate that flow occurs via the formation of transient shearbands; however, whether these flow inhomogeneities, which are also observed in steady flow [17–20], are consequences of the above-mentioned correlated processes remains to be resolved [18, 21, 22]. Also, recent stress-controlled granular experiments [23], while observing such shearbands, report the significance of local plastic events leading to avalanches and eventual flow [24]. So the question is whether these features are generic to the fluidization of amorphous systems under an imposed stress.

In this Letter, we report a simulational study of the onset of flow in a confined colloidal glass, under an imposed uniform shear stress. We use a geometry that mimics the typical planar Couette setup in stress-controlled experiments of soft materials. We show that indeed the timescale for the onset of steady flow diverges with decreasing stress, along with the emergence of nonlinear creep which has a power law form (similar to a range of materials [5, 7, 25–28]). Moreover, by following the local dynamics, we observe that the mobility is spatio-temporally heterogeneous and the mobile regions take the shape of shear-band-like

structures. Thus, our work provides evidence that in the presence of an external shear stress, the fluidization of the glass near yielding is extremely slow, along with the presence of long-lived spatial heterogeneities.

For our simulations, the model colloidal system that we consider is a 50 : 50 binary Yukawa fluid [29, 30]. Thus, interactions between the particles are given by the pair potential $V_{\alpha\beta}(r) = \varepsilon_{\alpha\beta} d_{\alpha\beta} \exp[-\kappa(r - d_{\alpha\beta})]/r$, with r the distance between a particle of type α and one of type β ($\alpha, \beta = A, B$). For model parameters, see [31]. Our MD simulations have been done for samples having the dimensions $L_x = 26.66d_s, L_y = 53.32d_s, L_z = 13.33d_s$, consisting of $N = 12800$ particles. We work in the NVT ensemble and the temperature is controlled by using a Lowe thermostat [32]. We equilibrate the system, using periodic boundary conditions, at a high temperature of $T = 0.2$ and then $m = 24$ independent configurations sampled at $T = 0.2$ were instantaneously quenched to $T = 0.05$ (which is below the mode-coupling critical temperature of $T_c = 0.14$ [29]). Subsequently, at $T = 0.05$, each of these m configurations are aged for durations of $t_{\text{age}} = 10^3, 10^4, 10^5$. When each configuration reaches a certain t_{age} , we freeze the particles at $0 < y < 2d_s$ and $L_y - 2d_s < y < L_y$ to prepare glassy states confined between rough walls. Then, these confined samples are sheared by pulling the top plate in $+x$ direction with a fixed force F_0 (similar to rheometers) [33], which imposes a constant shear stress of $\sigma_0 = F_0/L_x L_z$; we study the response of the confined glass to such an imposed field. Alternatively, a constant shear-rate ($\dot{\gamma}_0$) can be imposed by pulling the top plate at a constant velocity $v_0 = \dot{\gamma}_0(L_y - 4d_s)$ [20]; we used such a setup to estimate the dynamical yield stress (σ_d) of the confined glass at $T = 0.05$ [34], obtaining a value of $\sigma_d = 0.0857$.

Similar to experimental creep measurements, we study the response of the glass by monitoring the strain experienced at the top wall $\gamma_w(t)$. This is done by recording, for each of the m samples, the velocity of the wall $v_w(t)$ as a function of time, and then the average effective strain at the wall is obtained: $\gamma_w(t) = \langle \gamma_\alpha(t) \rangle_e$, where $\gamma_\alpha(t) = \int_0^t [v_w(t)/w] dt$ is the wall strain computed for α -th sample and $\langle \rangle_e$ is an average over the ensemble of m trajectories. In Fig. 1(a), we show how $\gamma_w(t)$ evolves for a wide range of imposed stress σ_0 . These curves show several regimes - (i) in all cases, at early times, the strain increases initially and then there is an oscillatory part, corresponding to the regime when the stress builds up inside the confined sample - the oscillations occur due to the interplay of the imposed stress at the wall and the restoring force of the deformed glass; (ii) after this, for large values of

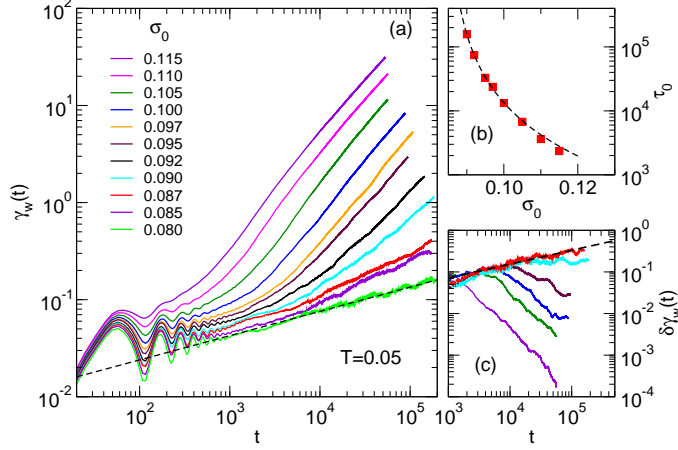


FIG. 1: (a) At an age of $t_{age} = 10^4$, evolution of strain at wall, $\gamma_w(t)$ for a range of imposed stress σ_0 . (b) Variation of timescale for onset of steady flow, τ_0 , as a function of σ_0 . The dashed black line corresponds to a fit to the data with $A/(\sigma_0 - \sigma_s)^\beta$, with $\sigma_s = 0.0848$ and $\beta = 2.285$. (c) Variation of strain fluctuations across trajectories, $\delta\gamma_w(t)$ for different σ_0 . The dashed line corresponds to a power law fit $\delta\gamma_w(t) \sim t^{1/3}$.

σ_0 , one quickly sees an asymptotic linear regime in $\gamma_w(t)$, which corresponds to a steady flow at a fixed shear-rate; (iii) as σ_0 is decreased, an intermediate regime in $\gamma_w(t)$ emerges, which for small enough σ_0 has a power-law behaviour - for the creep at $\sigma_0 = 0.80$ (which is below the dynamic yield stress σ_d), we obtain a power-law exponent of 0.25. For imposed stresses larger than σ_d , one eventually observes the asymptotic linear regime. Now, one can define a time-scale for onset of flow, τ_0 , as the time required for the system to reach a strain of $\gamma_w(\tau_0) = 1$; using this definition, we calculate τ_0 for each σ_0 and plot it in Fig. 1(b). One can see that the timescales increase with decreasing stress and the data can be fitted with the function $A/(\sigma_0 - \sigma_s)^\beta$, with $\sigma_s = 0.0848$ ($\approx \sigma_d$, the estimated dynamical yield stress) and $\beta = 2.285$ (which is similar to experimental observations [5, 7]).

In supercooled liquids, diverging timescales are seen to be associated with increasing dynamical heterogeneities. For the onset of flow in glasses, we explore this possibility by studying the fluctuations in response within the ensemble of m samples having the same age; this is quantified by calculating $\delta\gamma_w(t) = \langle \gamma_\alpha^2(t) \rangle_e / \langle \gamma_\alpha(t) \rangle_e^2 - 1$. In Fig. 1(c), we plot $\delta\gamma_w(t)$ for different σ_0 . We see that for large values of σ_0 , the function $\delta\gamma_w(t)$ increases with time, has a maximum and then decays at long times. The location of the maximum shifts to larger timescales with decreasing σ_0 , similar to the trend for τ_0 . As we approach the

yielding regime, the location of this maximum goes beyond our time window of observation. Interestingly, the initial increase in $\delta\gamma_w(t)$ has a power-law behaviour, which is identical for all σ_0 , as demonstrated in Fig. 1(c). This is similar to what was earlier observed in creep flow in experiments using paper samples [28] - a power-law in average strain, as well as a power law in the fluctuations.

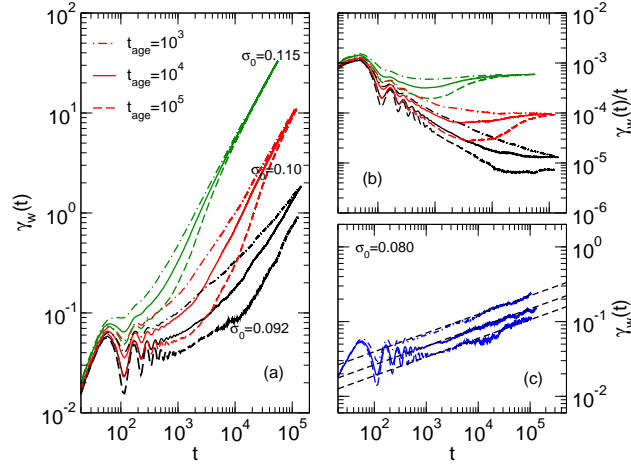


FIG. 2: (a) Wall-strain $\gamma_w(t)$ for samples having different ages ($t_{\text{age}} = 10^3, 10^4, 10^5$) for different imposed stresses $\sigma_0 = 0.115, 0.100, 0.092$. (b) Corresponding effective strain-rates, $\gamma_w(t)/t$, at the wall. (c) $\gamma_w(t)$, for $\sigma_0 = 0.080$ for the three ages, revealing identical power-law creep (with exponent of 0.25, indicated with dashed lines) in this sub-yielding regime.

Aging is a characteristic property of glass-forming systems around the glass-transition temperature T_g . Below this temperature, the transient response to a deformation does depend on the history of preparation [33, 35]. This has also been observed in the recent creep experiment on colloidal glass [7], where the deformation of samples having different ages was studied. In our simulations, the ages that we can explore are relatively smaller compared to experiments; despite that, we explore whether similar aging-dependence is also observed in our case. In Fig. 2(a), for different values of imposed stress $\sigma_0 = 0.092, 0.100, 0.115$, we plot the wall-strain $\gamma_w(t)$ for samples having different ages, viz. $t_{\text{age}} = 10^3, 10^4, 10^5$. The corresponding effective strain-rates at the wall, $\gamma_w(t)/t$, are shown in Fig. 2(b). As observed in the experiments, for $\sigma_0 = 0.100, 0.115$, we recover the same asymptotic regime for $\gamma_w(t)$, i.e. the final shear-rate is the same for all the aged samples, which is expected. However, the transient regime depends upon the age - the larger the age, the later it reaches the asymptotic linear regime with variation in the shape of $\gamma_w(t)/t$ when the system “breaks”

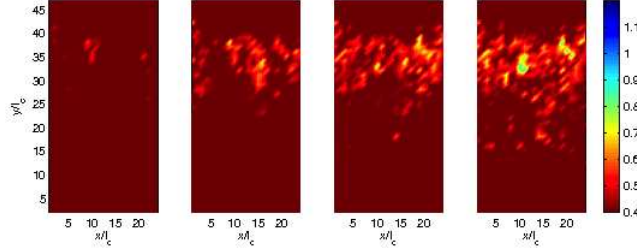


FIG. 3: For $\sigma = 0.080$, spatial maps of average transverse displacements along a single trajectory ($t_{\text{age}} = 10^4$). Measurements done at $t = 10790, 25730, 92960, 251905$ (from left to right), with the colorbar giving the scale of local mobilities.

into flow. In fact, the curvature in $\gamma_w(t)/t$ can also be different, which we observe for $\sigma_0 = 0.10$ (see Fig. 2(b)) - while the curve for $t_{\text{age}} = 10^5$ has a pronounced minimum, it is a monotonically decreasing function for $t_{\text{age}} = 10^3$. This implies that the intermediate flow states could be different for samples having different ages. For the case of $\sigma_0 = 0.092$, $\gamma_w(t)$ for $t_{\text{age}} = 10^5$ does not reach the steady-state within our window of observation, unlike the cases of $t_{\text{age}} = 10^3, 10^4$. In Fig. 2(c), we study the situation for the case when $\sigma_0 < \sigma_d$; for all the three different ages, at $\sigma_0 = 0.080$, $\gamma_w(t)$ has a power-law creep, with an exponent of 0.25. This indicates that the origin of the power-law behaviour is due to processes which are age-independent.

Until now, we have measured the macroscopic response by monitoring the motion of the top wall. We observed, with decreasing stress, the appearance of creeping dynamics. We now want to see how this slow deformation shapes up at a more local scale within the sample; whether the local response is uniform or heterogeneous and whether any spatial structures are formed as the system yields. In order to study this, we consider non-affine motions occurring due to local structural rearrangements. To this end, maps of transverse displacements are computed in the following manner. Before we apply the external stress ($t = 0$), we divide the xy plane of the simulation box into small square cells (of length $l_c = 1.1d_s$) and identify the particles in each cell. Next, after time t , we determine the transverse displacement of each particle $\Delta y_i(t) = |y_i(t) - y_i(0)|$ and then, to construct the spatial maps, we calculate for each cell the local mobility, $\mu_{lm}(t) = \langle \Delta y_i(t) \rangle_{lm}$ (with $\langle \rangle_{lm}$ the average over all the particles in the cell $\{lm\}$ at $t = 0$).

In Fig. 3, for a single trajectory of a sample aged to $t_{\text{age}} = 10^4$, we show the displacement

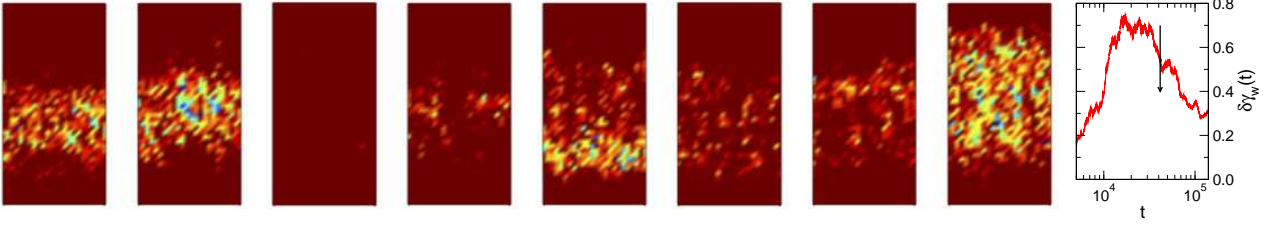


FIG. 4: For samples at $t_{\text{age}} = 10^5$, (left to right), the first eight panels show displacement maps for different trajectories at a fixed time $t = 42745$ for $\sigma_0 = 0.092$, indicating the variation in response of different samples at same age. (Last panel) $\delta\gamma_w(t)$ for the corresponding ensemble, with the arrow indicating the time of measurement.

maps as the system evolves in time, at an imposed stress of $\sigma_0 = 0.080$ (i.e. where we observe the power-law creep). Note that the local non-affine dynamics is extremely slow; even at $t = 10790$ (first panel on the left), only a few faint spots of significant displacements ($\mu_{lm}(t) > 0.5d_s$) are seen. As time progresses, these spots expand into larger patches ($t = 25730$). By $t = 92960$, we see that these patches form a band-like structure extending across the length of the box and this structure remains persistent till the end of our observation ($t = 251905$). In the rest of the system, even at such long timescales, the mobility is comparably negligible. Thus, during creep flow, while initial mobile spots seem to initiate activities in the neighbouring region along the flow direction, similar mobilities in the transverse direction seem to be impeded, which makes these localized structures persistent. For larger stresses where eventually steady-state flow is obtained, such spatial heterogeneities also occur; but they are temporally short-lived, with the mobile regions quickly building up transversely across the sample until the entire system is fluidized. Hence, the divergence of timescales for the onset of steady-state, with decreasing σ_0 , is probably associated with a slow emergence of regions of mobility as well as the impediment of propagation of flow in the transverse direction.

The displacement maps can now be used to clarify the origin of the temporal behaviour of the fluctuations in wall strain $\delta\gamma_w(t)$. For samples aged to $t_{\text{age}} = 10^5$, we study the dynamics for the independent trajectories evolving for $\sigma_0 = 0.092$. In Fig. 4(a), eight maps from this ensemble, calculated at $t = 42745$, are shown; the time of measurement is marked by an arrow in Fig. 4(b), which shows $\delta\gamma_w(t)$ for the entire ensemble. From map-to-map, one clearly sees that the local mobility is spatially different, with the occurrence of varying

degrees of localization (or even the absence of it). Thus, this implies that even though all these confined samples have the same age, the local response to an imposed stress differs from one to the other, depending upon the initial state. The varying degree of fluidization, within an ensemble, indicates a distribution of timescales for complete fluidization of each initial state; and this variation is captured within $\delta\gamma_w(t)$. Therefore, the increase in $\delta\gamma_w(t)$ with decreasing σ_0 implies that this distribution of timescales becomes broader as we approach the yielding regime.

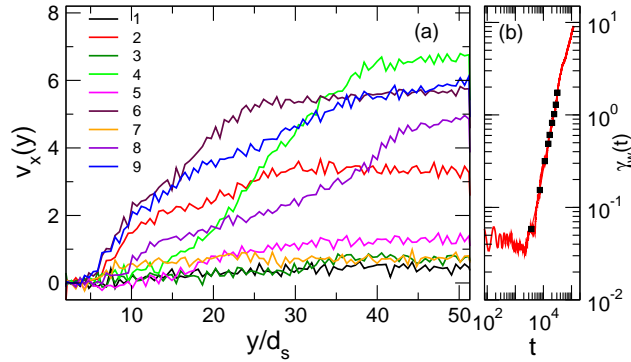


FIG. 5: (a) Velocity profiles during the onset of flow at $\sigma_0 = 0.10$ for a sample with age $t_{\text{age}} = 10^5$, averaged for $\Delta t = 498$, starting from different time-origins t_0 (labelled in sequence). (b) For the same trajectory, $\gamma_w(t)$ with the black squares marking the different t_0 .

The displacement maps, which track motion transverse to the flow direction, indicate the presence of heterogeneous dynamics; one can expect that the corresponding velocity profiles (which monitor the spatial dependence of the motion along the force direction) would also capture such heterogeneities. Recently, there has been focus on the possible occurrence of transient shear-bands [5, 36] in velocity profiles during the onset of flow in soft glasses. For a single trajectory at $t_{\text{age}} = 10^5$, we show such velocity profiles in Fig. 5(a), measured during flow at $\sigma_0 = 0.10$ (a case where one attains a steady-state at long times). Since instantaneous profiles are too noisy, we average velocity profiles over short intervals in time ($\Delta t = 498$) starting at different points in time (t_0) after the imposition of the stress. In Fig. 5(b), we plot the evolution of the wall-strain $\gamma_w(t)$ for this trajectory and the different t_0 are marked on the curve; they correspond to the timescales when the material “breaks” into flow. We see that, during these times, the velocity profiles fluctuate quite a lot, with signatures of intermittent flow (i.e. existence of little or no flow at some time instances, interspersed with burst of flow) as well as the existence of band-like profiles (co-existence of regions of no-flow

with more mobile regions). These transient heterogeneities in the velocity profiles, while in agreement with the observation in gels [5], seem to reflect a more complex motion in flow direction.

Creep flow observed in crystalline materials, are often explained via the motion of dislocations [37]; however, for the disordered materials, such processes are absent. For soft materials, different rheological models like SGR [38], phenomenological fluidity models [39], and schematic models [7] do predict creep flows of various forms; but neither do they take into account any spatial information for building the model, nor do they provide predictions for the local dynamics. In that aspect, the non-local fluidity model [11] seems promising; however, the temporal evolution of spatial patterns of local fluidity, as the material starts flowing, is yet to be worked out. Only recently, a more spatial version of the SGR model was used to show that creep flow is associated with shearbanded velocity profiles [40]. In this context, our simulations provide further insight into the possible ingredients for constructing a more complete theoretical model which would correctly describe the dynamics near yielding.

In conclusion, using numerical simulations, we have demonstrated that during the onset of Couette flow in glassy systems, creep (characterized by a power-law dependence of strain on time) is associated with strong spatial heterogeneities in the dynamics which take the form of long-lived shearbands. Moreover, for the confined system, the local response also depends on the initial structure, showing up as fluctuations within an ensemble. Thus, even for a simple planar Couette flow geometry, one observes a rich spatio-temporal response of the soft glass as it yields. Further studies are necessary to clarify how this transient behaviour connects to creep observed in uniaxial tensile studies of metallic [41] and polymeric [42–44] glasses. For soft amorphous materials, while more local measurements are necessary from experiments, simulations can also further explore yielding in different complex geometries.

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